

# Traffic-induced multicomponent ultrafine particle microphysics in the WRF v3.6.1 large eddy simulation model

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# **Traffic-induced multicomponent ultrafine particle microphysics in the WRF v3.6.1 large eddy simulation model: General behaviour from idealised scenarios at the neighbourhood-scale**

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25 **HIGHLIGHTS**

- 26
- 27 1) Multicomponent microphysics of UFPs is coupled with the WRF-LES model.
- 28
- 29 2) General behaviour of UFPs on the neighbourhood scale dispersion is investigated.
- 30
- 31 3) The combined effects of emissions, mixing and microphysics of UFPs are revealed.
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- 34

## 35    **ABSTRACT**

36    Traffic is the key source of ultrafine particles (UFPs, particulate matter with an aerodynamic diameter  
37    less than 0.1  $\mu\text{m}$  or 100 nm) in most urban areas. The traffic-generated UFPs vented out from an  
38    urban street mix with overlying ‘urban background air’ and are diluted whilst also undergoing change  
39    due to condensation/evaporation and other aerosol microphysics. Traffic-generated UFPs are  
40    comprised of a complex mixture of semi-volatile compounds (SVOCs) with volatility varying over  
41    many orders of magnitude, resulting in size-dependent particle composition. This study coupled the  
42    multicomponent microphysics (involving condensation/evaporation) of UFPs with the WRF v3.6.1  
43    (Weather Research and Forecasting) large eddy simulation model (i.e. WRF-LES-UFP), and used  
44    this modelling system to investigate the general behaviour of UFPs on the neighbourhood scale (10-  
45    1,000 m; transport times of few minutes) for idealised scenarios. The model captures the horizontal  
46    dispersion of UFPs downwind into the neighbourhood scale and vertical mixing with urban  
47    background air. Evaporation decreases the mode size of UFPs venting into the urban boundary layer  
48    from street-level. The neighbourhood-scale evolution of UFPs is, therefore, a combination of the  
49    effects of emissions, mixing with background, and condensation/evaporation. Total UFP number  
50    concentration and total mass concentrations scale linearly with the emission rate or the background  
51    concentration, demonstrating numerical conservation of the scheme. The linearity is less pronounced  
52    for the number concentration of smaller particles (UFP diameter less than 100 nm) with respect to  
53    UFP size and concentrations of those carbon components with a time scale comparable to the dilution  
54    time scale (in the order of minutes), reflecting the effects (altering the particle sizes) due to  
55    condensation/evaporation.

56

57    **Keywords:** Atmospheric nanoparticles; Urban pollution; Aerosol microphysics; Urban Street; Semi-  
58    volatiles.

59

## 1. INTRODUCTION

Ultrafine particles (UFPs or  $PM_{0.1}$ , particulate matter with an aerodynamic diameter  $D_p < 0.1 \mu m$ ) are respirable (Manigrasso et al., 2017) and may cause adverse health effects to the pulmonary system, and the cardiovascular/nervous systems (e.g. Panis et al., 2010; Geiser et al., 2005). Unlike larger size fractions  $PM_{10}$  ( $D_p < 10 \mu m$ ) and  $PM_{2.5}$  ( $D_p < 2.5 \mu m$ ) (US EPA, 2017; European Commission, 2017), there are currently no ambient air quality regulations for UFPs. UFPs dominate particle size number concentrations (Harrison et al., 2000) and have received increasing attention from the scientific community (e.g. Dall'Osto et al., 2011; Harrison et al., 2011; Vu et al., 2017; Jacobson et al., 2005). UFPs from vehicle emissions, which tend to dominate the urban atmosphere (Harrison et al., 2018; Kumar et al., 2014), contain multi-components of semi-volatile compounds (SVOCs) (Alam et al., 2016; Baldauf et al., 2016), contributing to the changes in particle size due to condensation/evaporation (Harrison et al., 2016; Jacobson et al., 2005).

Condensation and evaporation are among the most important aerosol microphysical processes in predicting the fate of ultrafine particles in urban air (Gelbard and Seinfeld, 1980; Harrison et al., 2016; Jacobson, 2005; Jacobson and Turco, 1995; Jacobson et al., 1996; Pankow, 1994). Nikolova et al. (2016) developed a CiTTy-Street-UFP box model, an urban version of CiTTyCAT Lagrangian model (Pugh et al., 2012) including aerosol microphysics. The model was used to assess the importance of microphysical processes and to simulate the behaviour of traffic-related UFPs within a street canyon and UFP evolution from canyon rooftop to a nearby downwind urban park. For a steady-state simulation of number concentration in the canyon, there is a balance among the traffic-related UFP emissions, aerosol microphysics, and exchange with the rooftop air. There is also evidence of the evaporation of UFPs when the rooftop air is advected to the neighbourhood park. Nikolova et al. (2018) further used this box model approach to investigate the influence of particle composition on the evolution of particle size distributions at the time scale of 100s (related to the dispersion at the neighbourhood scale). The evaporative shrinkage of UFPs from the nucleation mode

was highly influenced by SVOC composition. Zhong et al. (2018) coupled the UFP multicomponent microphysics (i.e. evaporation/condensation of SVOCs) with a two-box model for urban street canyon compartments and investigated factors that may inhibit mixing. The contrasts in the UFP number-size distribution between the lower and upper canyons are captured by this two-box-UFP canyon model. The traditional assumption of a one box model for a compartmentalised canyon would typically under-predict the UFP number concentration in the lower canyon compared with the proposed two-box model. The traffic-generated nanoparticles vented out from an urban street network, which may be simulated by a street canyon box model (Nikolova et al., 2016), could be evaporated to smaller particles while they are transported during neighbourhood scale dispersion (Dall'Osto et al., 2011; Harrison et al., 2019).

According to the horizontal length scale, Britter and Hanna (2003) classified the atmospheric flow and related phenomena (e.g. dispersion of pollutants) into four categories: i.e. regional scale (~100-200 km), urban/city scale (~10-20 km), neighbourhood scale (~1-2 km) and local/street scale (~0.1-0.2 km). The neighbourhood scale has been increasingly concentrated on by the urban research community. This is a scale over which the computation can be feasible at high spatial and temporal resolution (Carpentieri et al., 2012).

The large-eddy simulation (LES) mode of the Weather Research and Forecasting (WRF) model (WRF-LES) (Skamarock and Klemp, 2008) is a powerful tool to simulate the turbulence-resolved atmospheric flow at the neighbourhood scale in 3-dimensional (3D) Eulerian grids with high spatial and temporal resolutions. Nottrott et al. (2014) investigated the dispersion of a passive scalar from continuous point sources within the atmospheric boundary layer using the WRF-LES. The plume trajectories were captured by the model. Nunalee et al. (2014) also simulated the plume impingement of a passive scalar in the atmospheric turbulent flow influenced by complex terrain features. The spatial pattern of the surface plume trajectory was well revealed by the WRF-LES. Jacobson and

112 Seinfeld (2004) investigated the dispersion and the evolution of soot particle size distributions from  
113 both point and line sources using a 3D global-through-urban atmospheric model. There was a  
114 reduction in the particle number concentrations downwind of the emission sources, primarily due to  
115 dilution. The WRF-CHEM mesoscale model (Grell et al., 2005) provides the capability of coupling  
116 WRF with “online” chemistry involving several aerosol schemes, such as GOCART (From the  
117 Goddard Chemistry Aerosol Radiation and Transport model) (Chin et al., 2000), MOSAIC (Model  
118 for Simulating Aerosol Interactions and Chemistry) (Zaveri et al., 2008) and MADE/SORGAM (The  
119 Modal Aerosol Dynamics Model for Europe with secondary organic aerosols) (Ackermann et al.,  
120 1998; Schell et al., 2001). However, there is no aerosol size information for GOCART, only 4 or 8  
121 size bins up to  $PM_{10}$  for MOSAIC and 3 log-normal aerosol modes (i.e. Aitken, accumulation and  
122 coarse) for MADE/SORGAM. There are currently no prior studies using WRF-LES at the  
123 neighbourhood/micro scale and multicomponent aerosol microphysics for UFPs at the nanoparticle  
124 scale.

126 In this study, the multicomponent microphysics (i.e. condensation/evaporation) of UFPs is coupled  
127 with WRF-LES (WRF-LES-UFP) to simulate the evolution and dispersion of UFPs at the  
128 neighbourhood scale for idealised scenarios of road emissions. The UFP code was previously  
129 implemented into CiTTY-Street-UFP (Nikolova et al., 2016) and the compartmentalised canyon box  
130 model (Zhong et al., 2018). Condensation and evaporation in the presence of semi-volatiles are  
131 processes that can actively re-partition semi-volatiles between gas and particle, alter the sizes of the  
132 UFPs and change the chemical composition of UFPs. With the proposed model (WRF-LES-UFP),  
133 we can explore a very complex system of interactions between particles, emissions, atmosphere and  
134 mixing, and quantify those interactions.

## METHODS

### 2.1 The WRF-LES Model

The WRF-LES model explicitly calculates the larger resolved eddies with the smaller unresolved eddies parameterised by subgrid-scale (SGS) turbulence models (Moeng et al., 2007) and can be used for the neighbourhood scale simulation. LES simulations, therefore, allow for intermittency in turbulence in a way that cannot be captured by Reynolds-average Navier-Stokes (RANS) (e.g. Solazzo et al., 2008; Baik et al., 2007; Kwak and Baik, 2014) or semi-analytical Gaussian dispersion methods (e.g. McHugh et al., 1997; Popoola et al., 2018; Munir and Habeebullah, 2018). The filtered continuity and Navier-Stokes equations can be described as follows (Munoz-Esparza et al., 2015):

$$\frac{\partial \tilde{u}_i}{\partial x_i} = 0, \quad (1)$$

$$\frac{\partial \tilde{u}_i}{\partial t} + \frac{\partial \tilde{u}_i \tilde{u}_j}{\partial x_j} = \nu \frac{\partial^2 \tilde{u}_i}{\partial x_j \partial x_j} - \frac{1}{\tilde{\rho}} \frac{\partial \tilde{p}}{\partial x_i} - \frac{\partial \tau_{ij}}{\partial x_j} + f_c \epsilon_{ij3} (\tilde{u}_j - U_{g,j}), \quad (2)$$

where  $\tilde{u}_i$  represents the  $i^{\text{th}}$  component of resolved velocity with  $i$  (or  $j$ ) = 1, 2, 3 indicating x, y, z directions;  $x_i$  (or  $x_j$ ) denotes the  $i^{\text{th}}$  (or  $j^{\text{th}}$ ) component of spatial coordinates;  $t$  is time;  $\tilde{p}$  is the resolved pressure;  $\tilde{\rho}$  is the resolved density;  $f_c$  denotes the Coriolis parameter due to the rotation of Earth (here,  $f_c = 1.139 \times 10^{-4} \text{ s}^{-1}$ , representing a latitude of 51.526 N);  $\epsilon_{ij3}$  denotes the alternating unit tensor;  $U_{g,j}$  denotes the geostrophic wind;  $\nu$  is the kinematic molecular viscosity;  $\tau_{ij}$  denotes the SGS stress tensor, which is parameterised by the 1.5-order turbulent kinetic energy (TKE) SGS model (See Section A in the Supplementary Material for details).

### 2.2 Size-Dependent Multicomponent Microphysics of UFPs

The multicomponent microphysics of UFPs considered in the current WRF-LES model include the condensation and evaporation processes of SVOCs, which are the dominant microphysical processes in predicting the neighbourhood scale evolution of UFPs in urban air (Nikolova et al., 2016). For the purpose of this study, coagulation and deposition processes have been omitted as these play a far lesser role than evaporation processes on the short timescales of neighbourhood atmospheric transport



162 (Nikolova et al., 2016). Nikolova et al. (2016) suggested that the exclusion of coagulation (with a  
 163 coagulation kernel accounting for Brownian motion) and deposition processes in the street canyon  
 164 model would lead to a reduction of the total particle number by 4.7% - 8.2% depending on the  
 165 ventilation conditions. Jacobson et al. (2005) suggested that Brownian motion together with van der  
 166 Waals/viscous forces and fractal geometry were the three most important processes in the treatment  
 167 of coagulation kernel and it was essential to consider both evaporation and coagulation in the model  
 168 for nanoparticles below 15 nm near a roadway, especially when there was a peak diameter less than  
 169 10 nm for the nucleation mode. In this current modelling study, we focus on the relative importance  
 170 of the processes, i.e. mixing vs. condensation and evaporation processes, emission vs. background.  
 171 By using the sectional modelling approach, the mass transfer rate between particulate and gas phases  
 172 due to the condensation and evaporation processes for the  $q$ -th component SVOC of one particle in  
 173 the  $jb$ -th size bin can be estimated as follows (Jacobson, 2005):

$$\frac{d\tilde{m}_{q,jb}}{dt} = a_{FS}^{jb} \frac{2\pi D_{p,jb} M_q D_q}{RT} (e_q^\infty - X_{q,jb} a_K^{q,jb} e_q^{vap}), \quad (3)$$

175 where  $q$  is for the  $q$ -th component;  $jb$  is for  $jb$ -th size bin;  $a_{FS}$  is the Fuchs-Sutugin correction factor;  
 176  $D_p$  represents the particle diameter (m);  $M$  is the molar mass ( $\text{g mol}^{-1}$ );  $D_q$  denotes the vapour  
 177 diffusivity ( $\text{m}^2 \text{s}^{-1}$ );  $R$  is the universal gas constant ( $\text{J mol}^{-1} \text{K}^{-1}$ );  $T$  is the temperature (K);  $e_q^\infty$  is the  
 178 partial pressure in the ambient air (Pa);  $X$  is the molar fraction;  $a_K$  accounts for the Kelvin effect;  
 179  $e_q^{vap}$  is the saturation vapour pressure (Pa), i.e. the vapour pressure of the chemical species  
 180 evaporating from the particle surfaces.  $e_q^\infty$  is derived from the gas-phase concentration of component  
 181  $q$  based on the ideal gas law, while  $e_q^{vap}$  is based on the method of (Compernelle et al., 2011) found  
 182 in the UManSysProp online tool (Topping et al., 2016). The saturation vapour pressure using the  
 183 method of Compernelle et al. (2011) was tested and compared with other vapour pressure estimation  
 184 methods in Nikolova et al. (2018) and informed by measurements of vapour pressure made in our  
 185 laboratory (Alam et al., 2019).

Table 1 shows the initial partial pressure at the inlet of the model and the saturation vapour pressure estimated at temperature of 284.15 K based on the UManSysProp online tool using data from Compennolle et al. (2011). When  $\frac{d\tilde{m}_{q,jb}}{dt} > 0$  in Equation 3, vapour condenses on the particles; when  $\frac{d\tilde{m}_{q,jb}}{dt} < 0$  in Equation 3, evaporation of SVOCs from the particle phase takes place. The overall production of gaseous component  $q$  due to multicomponent microphysics of UFPs can be calculated as:

$$\Delta\tilde{c}_q = -\sum_{jb} \tilde{N}_{jb} \frac{d\tilde{m}_{q,jb}}{dt}, \quad (4)$$

where  $\tilde{N}_{jb}$  is the number concentration for the  $jb$ -th size bin. A dynamical size ( $d_{jn}$ ) is calculated for each size bin after the condensation and evaporation processes and a redistribution scheme is implemented to redistribute both UFP number and mass concentrations to the sectional bin (See Section B in the Supplementary Material for details).

The UFP composition in our default UFP module configuration includes 18 components, i.e. 1 non-volatile core and 17 surrogate n-alkane components (SVOCs), i.e. C<sub>16</sub>H<sub>34</sub>-C<sub>32</sub>H<sub>66</sub> (labelled ‘C16’ – ‘C32’, below). The hundreds of SVOC components found in UFP (Alam et al., 2016) are mapped onto these surrogates on the basis of their volatility (Nikolova et al., 2018) (See Table S1 in the Supplementary Material for the volatility bin information, grouped n-alkane/surrogate n-alkane used in this study). These volatility classes are derived based on the GC × GC chromatogram with the same carbon numbers in different carbon arrangements (e.g. straight-chain or branched-chain) (Alam et al., 2016). Particle size is sectioned into 15 equally-sized bins on a logarithmic scale, covering particles with geometric mean diameter of 6.7 nm – 501.4 nm. There are 17 SVOC components in the gas-phase, corresponding to each surrogate n-alkane. The UFP number concentration of each size bin is updated diagnostically based on the total mass concentration of the size bin.

### 2.3 Framework of WRF-LES-UFP Coupling

The dispersion of tracers (for both multicomponent particulate and gas phases) in the WRF-LES model is described as follows. For the multicomponent particulate phase:

$$\frac{\partial \tilde{Q}_{q,jb}}{\partial t} + \frac{\partial}{\partial x_j} (\tilde{Q}_{q,jb} \tilde{u}_j) = \frac{\partial}{\partial x_j} \left( K_c \frac{\partial \tilde{Q}_{q,jb}}{\partial x_j} \right) + \Delta \tilde{Q}_{q,jb} + E_{q,jb}, \quad (5)$$

where  $\tilde{Q}$  represents the resolved UFP mass concentration; “ $q$ ” denotes the  $q$ -th component; “ $jb$ ” denotes the  $jb$ -th size bin;  $K_c$  is the SGS eddy diffusivity;  $\Delta \tilde{Q}$  and  $E_{q,jb}$  represent the resolved source terms due to multicomponent microphysics of UFPs (i.e. the condensation/evaporation processes included in the current study) and emissions, respectively.

For the multicomponent gas phase,

$$\frac{\partial \tilde{c}_q}{\partial t} + \frac{\partial}{\partial x_j} (\tilde{c}_q \tilde{u}_j) = \frac{\partial}{\partial x_j} \left( K_c \frac{\partial \tilde{c}_q}{\partial x_j} \right) + \Delta \tilde{c}_q + E_q, \quad (6)$$

where  $\tilde{c}_q$  represents the resolved gas concentration for component  $q$ ;  $\Delta \tilde{c}_q$  and  $E_q$  represent its resolved source terms due to multicomponent microphysics of UFPs and emissions, respectively. The resolved source terms for both particulate and gas phases in Equations 5-6 are derived from the UFP multicomponent microphysics module (involving condensation/evaporation), as in previous canyon box models (Nikolova et al., 2016; Zhong et al., 2018; Nikolova et al., 2018).

### 2.4 WRF-LES-UFP Model Configuration and Scenarios

To demonstrate the capabilities of the WRF-LES-UFP model, we choose an idealised urban scenario driven by realistic environmental conditions and boundary conditions. Such an idealised scenario allows us to diagnose more readily the UFP dynamics. The WRF-LES computational domain used in this study (Figure S1 in the Supplementary Material) covered a horizontal area of  $2.54 \text{ km} \times 2.54 \text{ km}$  ( $L_x \times L_y$ ) with  $20 \text{ m} \times 20 \text{ m}$  resolution. The vertical domain size is  $1 \text{ km}$  ( $L_z$ ) with 79 stretch grids, so

the number of cells in the domain is  $127 \times 127 \times 79$ . Because the length scales of the underlying building geometries and street canyons are about same as the grid resolution adopted here (20 m), it is not appropriate to resolve these structures explicitly. It is assumed that the height and roughness of the canopy vary slowly relative to the grid resolution, and a homogeneous urban canopy is used in this study. To focus on the evolution of multicomponent UFPs at the neighbourhood scale above the urban canopy (instead of building scale inside the urban canopy), we adopt a simplified approach of setting up the first vertical cell at a nominal ‘rooftop level’ and specifying momentum flux and heat flux at the bottom of the domain using observational data as discussed below.

A fixed time step of 0.2 s is adopted for the airflow calculation, whilst an adaptive time step is used for solving multicomponent condensation/evaporation of UFPs. Periodic lateral boundary conditions are specified for velocity components. Inlet conditions for the SVOC quantities (gas concentrations, particle number concentration size distributions, and particle SVOC mass fractions) are specified as fixed values. At the outlet, a zero-gradient condition is specified for all SVOC quantities.

Weather conditions adopted for the simulations are informed by observational data (Heathrow airport) for the  $180^\circ \pm 25^\circ$  wind sector during the London campaign (Jan-Feb 2017) (see Harrison et al. (2019) for details). The geostrophic wind is specified as  $U_g = 5 \text{ m s}^{-1}$  and  $V_g = 10 \text{ m s}^{-1}$  and in the presence of the Coriolis force, this attempts to achieve an approximate southerly wind of  $2 \text{ m s}^{-1}$  near the rooftop ‘surface’ at the bottom of the model domain. An upwelling surface sensible heat flux of  $13 \text{ W m}^{-2}$  (tke\_heat\_flux in the WRF namelist.input option) is adopted for the lower boundary condition of heat and a surface drag coefficient of 0.048 (tke\_drag\_coefficient in the namelist.input option) is specified for the lower boundary condition of momentum, derived from eddy-covariance measurements at the rooftop of Regent's University during the campaign.

256 The boundary layer height is initially  $\sim 500$  m, specified by an initial constant vertical potential  
257 temperature of 282.15 K for the lower 475 m, capped by a strong inversion with of  $\partial T/\partial z = 0.05$  K  
258  $\text{m}^{-1}$  from 475 m to 625 m and  $\partial T/\partial z = 0.003$  K  $\text{m}^{-1}$  from 625 m to the domain top. The meteorological  
259 conditions of a low sensible heat flux at the ground and a strong capping inversion lead to a nearly  
260 constant height of the boundary layer during the simulation period. A damping layer of 300 m is  
261 applied near the domain top to prevent gravity waves. WRF-LES was run without the UFP module  
262 for 8 hours as a spin-up period in order to achieve a quasi-steady flow. Then the UFP module was  
263 switched on for 30 min, sufficiently long compared with the timescale of the geostrophic wind  
264 advection across the domain which is about 4 min. The output of the last 10 min with an interval of  
265 3 s was used for analysis.

266  
267 An idealised street (represented by a line emission perpendicular to the surface wind direction) with  
268 a width of 40m is configured in the middle of the domain (Figure S1 in the Supplementary Material).  
269 The emission fluxes to the first vertical model level is based on those from vehicles directly into the  
270 street canyon. In this study, we introduce a BASE case scenario and the settings are as follows. The  
271 emission factor for total UFP number is  $7.93 \times 10^{13}$  particles vehicle $^{-1}$  km $^{-1}$ , which is based on the  
272 value from Jones and Harrison (2006) with a correction factor for the year of 2017 derived from an  
273 analysis of measurement data (Harrison et al., 2019). The traffic activity is specified as 3740 vehicle  
274 hour $^{-1}$  at a heavily trafficked street (Marylebone Road) for the 180° wind sector during the campaign  
275 in London (Harrison et al., 2019).

276  
277 The size distribution of emitted UFPs is comprised of three log-normal distributions with peaks at 21  
278 nm, 29 nm and 69 nm, respectively, which are derived from the mode fitting for particle size  
279 distributions of the traffic increment, i.e. the background subtracted from Marylebone Road  
280 measurement at the 180° wind sector (Harrison et al., 2019). The mass fraction of non-volatile core

281 is assumed to be 1 % for the nucleation mode (Nikolova et al., 2016), gradually increasing to 90 %  
282 for the Aitken mode and 99 % for the accumulation mode. The mass fraction of SVOC compounds  
283 in the emitted UFPs is then scaled by the measured UFP composition in samples collected in  
284 Marylebone Road (Harrison et al., 2018).

285  
286 The emission factors of total grouped SVOCs (from C16 to C32) are  $681 \mu\text{g veh}^{-1} \text{ km}^{-1}$  for the gas  
287 phase and  $1714 \mu\text{g veh}^{-1} \text{ km}^{-1}$  for the particle phase (See details in Table S2 in the Supplementary  
288 Material). The UFP size distribution at the inlet of the domain is based on the mode fitting of BT  
289 tower measurements (160 m above ground level) for the  $180^\circ$  wind sector during the campaign in  
290 London (Harrison et al., 2019), which has 3 log-normal distributions with peaks at 24 nm, 66 nm and  
291 163 nm, respectively. The SVOC concentrations (gas and particles) at the inlet of the domain are  
292 based on the scaled Regent's Park measurements (inferred from the ratio of black carbon measured  
293 at BT tower to that measured at Regent's Park) (See Table S3 in the Supplementary Material for inlet  
294 SVOC concentrations). The inlet air parcel may be slightly adjusted by the multicomponent  
295 microphysics of UFPs to reach a quasi-equilibrium state after a certain travelling distance and we  
296 take 200 m before the emission at the bottom level of the domain (Figure S1 in the Supplementary  
297 Material) as the inflowing background in the analysis of the model output.

298  
299 In order to investigate the effect of emissions, cases with changes in emissions only for both gas and  
300 particle phases are configured (i.e., multiplying by a coefficient,  $\beta=[0.5, 0.75, 1.25, 1.5]$ , the  
301 emissions in the BASE case) and named EM0.5, EM0.75, EM1.25, and EM1.5, respectively. In order  
302 to investigate the effect of inlet background, cases with changes in inlet background only for both gas  
303 and particulate phases are configured using a coefficient  $\alpha=[0.5, 0.75, 1.25, 1.5]$ , i.e. case BG0.5,  
304 BG0.75, BG1.25, and BG1.5. We perform a linearity analysis by investigating the sensitivity to  
305 emissions and inlet background concentrations in both gas and particle phases.

306

### 307     **3.         RESULTS AND DISCUSSION**

#### 308     **3.1         The BASE Case Output from the model**

##### 309     **3.1.1       Total UFP number concentration**

310     Figure 1(a) illustrates a vertical slice of the total UFP number concentration (UFPNC), which is  
311     averaged horizontally in the cross-wind direction and temporally over the final 10 min simulation;  
312     this gives an indication of cross-sectional dispersion of UFPs for the BASE case in the model. The  
313     total UFPNC is gradually diluted by a southerly wind from the street (the west-east line emission) to  
314     its northerly neighbourhood. The vertical expansion of the plume is observed as UFPs are advected  
315     downwind into the neighbourhood scale and vertically mixed with urban background air.

316  
317     Similar neighbourhood dispersion behaviour was also observed by other simulations, i.e. Nottrott et  
318     al. (2014) and Nunalee et al. (2014) for the dispersion of a passive scalar from point sources and  
319     Jacobson and Seinfeld (2004) for the dispersion of soot particles from both point and line sources.  
320     The spatial pattern also indicates the neighbourhood dispersion of UFPs with a decrease of the total  
321     UFP number concentration downwind of the emission. This is consistent with classical line-source  
322     dispersion (Munir and Habeebullah, 2018; Jacobson and Seinfeld, 2004) and with the measured data  
323     which demonstrate that the UFPNCs in Marylebone Road were always much higher than those at its  
324     downwind neighbourhood rooftop site in Regent's Park (Dall'Osto et al., 2011; Harrison et al., 2019).  
325

326     Figure 1(b) shows a vertical slice of the ratio of concentration fluctuation intensity to mean  
327     concentration for total UFPNC for the BASE case in the model. The concentration fluctuation  
328     intensity is defined as  $C' = \sqrt{(C - C_m)^2}$ , where  $C_m$  represents a spatial and temporal average defined  
329     earlier. The value of  $C'/C_m$  can be interpreted as the percentage of particle number fluctuation  
330     intensity in relation to its mean value caused by the unsteady turbulence generated within the LES  
331     simulation. The region upwind of emission is more determined by the fixed inlet conditions and

therefore has no significant concentration fluctuation. Once emissions are released, there is a plume expansion for the concentration fluctuation intensity in the downwind side of the emission line.

Figure 1(c) illustrates the total UFPNC (both mean concentrations and concentration fluctuation intensities) along a South-North line at the bottom model level (corresponding to just above rooftop height) as a further indication of neighbourhood dilution. The highest value indicates the effect of emissions from the street, which is similar to Figure 7(a) in Jacobson and Seinfeld (2004) for the point source dispersion. The total UFPNC decreases rapidly at the near-source downwind neighbourhood, gradually approaching the background levels. This is due to the expansion of plume as it traverses its neighbourhood downwind (Figure 1(a)). In general, the total UFPNC appears to behave like a passive scalar, as expected from the model's number-conserving numerics and lack of (because assumed slow (Nikolova et al., 2016; 2018)) coagulation, deposition, and gas-phase chemistry.

### 3.1.2 UFP number-size distribution

Figure 2(a) illustrates the spatially (west-east) and temporally (the final 10 min period) averaged UFP number-size distribution (UFPNSD), together with its fluctuation intensity (indicated by shaded areas), for several downwind locations at the bottom level of the domain (corresponds to the urban canopy above rooftop height) for the BASE case in the model. The black line in Figure 2(a) represents the inflowing background (200 m before the emission at the bottom level of the domain). The red line with circles at the top of the graph is for the UFPNSD of the south-side rooftop cell of the line emission source. For comparison of mode sizes, the red line with triangles at the bottom is for UFP emission rate; other lines are for the UFPNSD at downwind distances of 100 m and 400 m (or travelling time of about 50 s and 200 s), respectively. Within each WRF-LES time step of 0.2 s, both south-side and north-side rooftop cells at the line emission source receive a given amount of emitted pollutants (both particle and gas phases) and the execution of WRF's advection and diffusion modules



358 brings some fresher air advected from the upwind neighbour cells. Then the UFP module is applied  
359 to the mixed air parcel and to yield a new UFPNSD for the south-side rooftop cell, for example, as  
360 shown by the top curve in Figure 2(a). Comparison of this curve with the emission curve gives a clear  
361 indication of the direct influence of emission, e.g. by their same peak diameter values (peaked at bin  
362 5 with bin bounds of [19.8 nm, 26.9 nm]). The small fluctuations of UFPNSD for the Aitken and  
363 accumulation modes (i.e. large size bins in Figure 2(a)) is explained by large fractions (90%-99%) of  
364 non-volatile core for this mode. An increase in the non-volatile core fraction will result in a decrease  
365 in the evaporative SVOCs (Nikolova et al., 2018).

366

367 There is a reduction in UFPNSD and fluctuations along the downwind distance in the simulation.  
368 There is also a clear shift of the peak particle size from the rooftop (peaked at bin 5 with bin bounds  
369 of [19.8 nm, 26.9 nm]) to its downwind locations of both 100m and 400 m (peaked at bin 4 with bin  
370 bounds of [14.6 nm, 19.8 nm]). This shift indicates particle evaporation during the neighbourhood  
371 dispersion of UFPs. The UFPNSD within the downwind neighbourhood is, therefore, a result of a  
372 combination of mixing and aerosol microphysics (i.e., condensation/evaporation of SVOCs). In the  
373 measurement in Harrison et al. (2019), there is also clear evidence of the shrinkage of particle sizes  
374 from the roadside (MR\_OBS in Figure 2a with a peak diameter falling into bin 5 [19.8 nm, 26.9 nm]  
375 as defined in the model) to nearby downwind rooftop location (RU\_OBS in Figure 2a with a peak  
376 diameter falling into bin 4 [14.6 nm, 19.8 nm] as defined in the model), with the lower diameter limits  
377 of 10 nm and 16 nm by the instruments adopted. This suggests that the current model can capture the  
378 general behaviour of particle size shrinkage mainly due to evaporation. The exclusion of coagulation  
379 process in our model may lead to an overestimation in the number concentration for particles below  
380 15 nm, as suggested by Jacobson et al. (2005) with a peak diameter less than 10 nm for the nucleation  
381 mode. Since the nucleation mode in our model has a peak diameter higher than 15 nm, the effect of  
382 coagulation process may be reduced. As there are many complex conditions in the real world  
383 measurement that our current model is not able to fully represent, such as well-defined background

384 conditions, emission pattern, real meteorological conditions together with realistic landscape,  
385 complex aerosol microphysics processes and other model input assumptions, it is difficult for us to  
386 well match the exact number size distribution profile measured in the field campaign.

387

388 Figure 2(b) illustrates the dilution ratio, defined as  $(C_{downwind} - C_{bg}) / (C_{rooftop} - C_{bg})$ , of UFP size  
389 fractions for downwind locations (rooftop+100m and rooftop+400m) for the BASE case in the model.  
390 For UFP size fractions with diameter greater than 100 nm, the dilution ratio closely follows that of a  
391 passive scalar (shown as size-independent dotted lines in Figure 2(b)). For these larger UFPs, the  
392 neighbourhood dispersion is dominated by dilution, attributable to the large fraction of non-volatile  
393 core in these particles. There are variations in dilution ratio for UFPs smaller than 100 nm, indicating  
394 that both dilution and condensation/evaporation are competing during the neighbourhood dispersion.  
395 Specifically, the number of medium-size particles ( $20 \text{ nm} < D_p < 100 \text{ nm}$ ) is reduced more quickly  
396 than the rate for a passive scalar, whereas the number of small-size particles ( $D_p < 20 \text{ nm}$ ) is reduced  
397 more slowly than the rate for a passive scalar. Because of the simplicity of our idealised scenario, we  
398 can attribute this size-dependent behaviour to evaporation, which converts some medium-size  
399 particles to smaller-sized ones during the advection-dilution process.

400

401

### 402 3.1.3 SVOCs mass concentrations

403 Figure 3(a-c) shows SVOC mass concentrations in both gas and particle phases at the rooftop and  
404 downwind neighbourhood locations for the BASE case in the model. For low-carbon components  
405 (C16-C18) having higher saturation vapour pressures (Table 1), the emissions are dominated by the  
406 gas phase (Table S2) and their respective rooftop gas concentration increments (from the background)  
407 are due to the vapour emissions. For the carbon components of C19-C22, the mass emissions are  
408 dominated by the particle phase (Table S2). However, their rooftop particle concentration increments

409 (Figure 3b) are not significant, particularly for C19-C21. On the other hand, their rooftop gas  
410 concentration increments are of large values (Figure 3a). This simulation clearly indicates that the  
411 evaporation process proceeds very rapidly to generate higher gas concentrations (Figure 3a) before  
412 the dispersion process takes effect. This can also be seen from the particle mass concentrations which  
413 are very close to background levels at the rooftop although they are subject to particle emission fluxes  
414 (Figure 3(b)). For C22, its rooftop particle concentration increment is about  $8 \text{ ng m}^{-3}$  (Figure 3(b)),  
415 whereas its gas concentration increment is about  $20 \text{ ng m}^{-3}$  (Figure 3a).

416

417 For carbon components of C23 onwards, the mass emissions are still dominated by the particle phase  
418 (Table S2), but their rooftop concentration increments are gradually shifted from gas phase to particle  
419 phase. The contribution of evaporation is important during the neighbourhood dispersion process for  
420 SVOCs having evaporation timescales of SVOCs (Table S4 in the Supplementary Material)  
421 comparable to the dilution time scale (i.e. for C21-C26) (Nikolova et al., 2018). This is indicated by  
422 the gas concentrations, especially for C24-C26, not decreasing appreciably during the neighbourhood  
423 dispersion.

424

425 For higher carbon components (from C27 onwards), condensation/evaporation is relatively slow due  
426 to their lower saturation vapour pressures (Table 1). The concentrations of these SVOCs in both gas  
427 and particle phases are more dependent on the strength of both emissions and background. As the gas  
428 emission fluxes are relatively smaller than particle, their gas-phase concentrations increase slightly  
429 at the rooftop and then approach background levels after a downwind distance of 100m. The  
430 increment due to emission is more significant in particle concentrations of these higher carbon  
431 number SVOCs. During the dispersion process, condensation processes may occur due to the very  
432 low saturation vapour pressures for some higher carbon components, e.g. C31-C32 (Table 1).

433

434 As shown in Figure 3(c), the rooftop generally has the highest concentrations (both mean and  
435 fluctuation concentrations) for all SVOCs (the sum of gas and particle phase concentrations)  
436 compared with the downwind locations. The SVOC concentrations rapidly decrease at a short  
437 travelling distance and then approach the background levels during their neighbourhood-scale  
438 dispersion. Because of mass conservation, each  $C_{n, \text{gas+particle}}$  should behave like a passive scalar,  
439 possessing the spatial patterns similar to total UFP number concentration shown in Figure 1, with  
440 their own background concentration and peak concentration level near the line source. Figure 3(d)  
441 illustrates the dilution ratio of SVOC gas and particle phases (defined in the same way as that for UFP  
442 number concentrations in Figure 2(b)) for the downwind locations (rooftop+100m and  
443 rooftop+400m). For both lower carbon gas-phase components (C16-C20) and higher carbon particle-  
444 phase components (C27-C32), the dilution ratio closely follows that of a passive scalar (as indicated  
445 by dashed lines, which are also the same as that in Figure 2(b)), indicating that dilution dominates  
446 during their neighbourhood dispersion.

447

448 For C21-C26, there are variations of dilution ratios along the dashed lines (for a passive scalar)  
449 indicating that aerosol condensation/evaporation plays an important role during the neighbourhood  
450 scale dispersion. Their gas-phase mass concentrations are reduced more slowly than the rate for a  
451 passive scalar (the dilution ratios are above the dashed lines) and their particle-phase mass  
452 concentrations are reduced faster than the rate for a passive scalar (the dilution ratios are below the  
453 dashed lines).

454

455 In summary, the above discussions suggest that the evaporation of low-carbon components (C16-  
456 C20) of emitted particles is almost complete on the timescale of less than a second (Table S4 in the  
457 Supplementary Material), and this leads to very high  $C_{\text{gas}}$  and negligible  $C_{\text{particle}}$  at the rooftop (Figs  
458 3(a) and 3(b)). From the rooftop location to the downwind locations of 100m and 400 m (or travelling

time of about 50 s and 200 s),  $C_{gas}$  for C16-C20 follows the dilution rate of a passive scalar (Figure 3(d)). However, the evaporation of the medium-carbon components (C21-C26) of emitted particles is slower, taking the timescale of travelling from the rooftop to 100-400 m downwind (about 50 s and 200 s). This evaporation process converts some medium-carbon component SVOCs from the particle phase to gas phase while the advection-dilution processes are underway during the period (Figure 3(d)). Finally the evaporation (or condensation) of high-carbon components (C27-C32) of emitted particles is extremely slow and within the timescale of travelling,  $C_{particle}$  behaves like the dilution of a passive scalar (Figure 3(d)).

467

### 468 **3.2 Linearity Analysis Among Cases from the model**

469 To provide insight into the relationships among multiple processes, the linearity of the dependence  
470 of several UFP quantities (UFPNSD, all-size particle mass concentration, gas mass concentration,  
471 and total mass concentration) on the emission rate and on the inlet background concentration, has  
472 been investigated. For this purpose, a scaled concentration for the  $ic^{th}$  case is defined as follows:

$$473 \quad \hat{C}^{ic} = (C^{ic} - \alpha C_{bg}^{BASE}) / \beta, \quad (7)$$

474 where  $C^{ic}$  is the concentration of any quantity in the  $ic^{th}$  case,  $C_{bg}^{BASE}$  is the background concentration  
475 in Case BASE,  $\alpha$  and  $\beta$  are the multiplied coefficients for inlet background and emission, respectively  
476 (defined in Sect. 2.3). If the linearity is perfect, the profile for any tested case (with the background  
477 subtracted) will scale exactly with total emission flux; in other words, the curves of scaled  
478 concentrations ( $\hat{C}^{ic}$ ) for all tested cases should collapse into a single curve. Variations in the curves  
479 of scaled concentrations,  $\hat{C}^{ic}$ , would indicate nonlinear behaviour of the UFP system. This test  
480 determines whether a reduced microphysical model can be adopted or not: for perfectly scaled  
481 behaviour, results for one case can be used to construct the solutions for any multiplication of the  
482 emission profile and/or the background concentration profile.

483

484 Figure 4 demonstrates the perfect scalability for the spatially and temporally averaged total UFPNC  
 485 along a South-North line at the bottom level of the domain in the model. All the tested cases, with  
 486 varying either  $\alpha$  or  $\beta$ , collapse onto a single curve, indicating an excellent scalability for total UFPNC.  
 487 This implies that if the results of a base case are known, the results for any other cases (corresponding  
 488 to a pair of  $\alpha$  and  $\beta$ ) can be obtained simply from the scaled equation (Equation 7), i.e.  $\hat{C}^{ic} =$   
 489  $\beta \hat{C}^{BASE} + \alpha C_{bg}^{BASE}$ .

490

491 For the rooftop location, there is weak non-linearity (not shown here) as the UFPNSDs are  
 492 predominately affected by emissions on a very short time scale (only influenced by lower carbon  
 493 components). Stronger non-linearity, for the downwind location of rooftop+400 m (travelling time of  
 494 about 200 s), is presented in Figure 5(a). The number concentrations for particles with diameters  
 495 higher than 100 nm exhibit a very good scalability. This is due to the large mass fraction of non-  
 496 volatile core in this size range, reducing the effects of evaporation/condensation, so that the behaviour  
 497 for these particles is very similar to that of passive scalars. For smaller particles, number  
 498 concentrations are not perfectly scaled. This is due to the nonlinear evaporation process for smaller  
 499 particles and the change of their particle sizes. If the scaled model (Equation 7) is used to generate an  
 500 approximate solution, then there would be an error for a non-perfectly scaled quantity (e.g. number  
 501 concentrations for smaller size bins). For the value range of  $\alpha$  and  $\beta$  tested here, the errors are not  
 502 remarkably large, as illustrated in Fig 5. In order to reveal the details of the scalability, a correction  
 503 factor defining the error for the  $ic^{th}$  case ( $\phi^{ic}$ ) relative to case BASE for the scaled quantity is,

$$504 \quad \phi^{ic} = (\hat{C}^{ic} - \hat{C}^{BASE}) / \hat{C}^{BASE}, \quad (8)$$

505

506 Figure 5(b) shows correction factors for selected nonlinear bins at the downwind location of  
 507 rooftop+400m in cases with varying background and emissions from the model. Correction factors

508 for selected bins are within 10% in most cases for varying emissions and varying background.  
509 Correction factors for bin 3 (with bin bounds of [10.7 nm, 14.6 nm]) are generally bigger than those  
510 for bin 7 ([36.6 nm, 49.9 nm]), followed by bin 5 ([19.8 nm, 26.9 nm]). For cases of varying  
511 background, bin 3 always has an opposite sign compared to bins 5 and 7. From the discussion  
512 associated with Figure 2(b) (the output of the BASE case), it is evident that the number concentration  
513 of bin 3 increases with the evaporation from particles of bins 5-9. Furthermore, the discussions  
514 associated with Figure 3(d) reveal that the evaporation occurs mainly to C21-C26 for a distance of  
515 100-400m (travelling time of about 50 s and 200 s). The decreasing trend of  $\phi$  for “BG-bin3” with  
516  $\alpha$  (indicator of the background concentrations’ magnitude) in Figure 5(b) suggests that higher  
517 background SVOC concentrations will suppress the evaporation processes of C21-C26 in bins 5-9,  
518 thus reducing particle numbers gained in bin 3. This finding is further supported by the increasing  
519 trend of  $\phi$  for “BG-bin7” with  $\alpha$  in Figure 5(b), as suppressed evaporation causes fewer particle  
520 numbers in bin 7 lost to smaller bins. It is interesting to see the trend of curve for “BG-bin5” is similar  
521 to that for “BG-bin7”, suggesting the particles of bin 5 also contribute to evaporation which is  
522 suppressed by higher background concentrations. This suppression of evaporation, however, is not  
523 clearly seen for the “EM” cases in Figure 5(b).

524

525 Figure 6 shows the scaled concentrations for both SVOC gas and particle concentrations at the  
526 downwind location of rooftop+400m (travelling time of about 200 s) from the model. There are very  
527 good scalabilities for lower carbon components (e.g. C16-C22). This may be attributed to their high  
528 vapour pressure and fast evaporation processes which would transfer particle SVOC mass to vapour  
529 very quickly (Nikolova et al., 2018). Then this would be an effective dilution process for gas  
530 concentrations (indicating nearly zero particle concentrations for lower carbon numbers in Figure  
531 6(b)).

532

533 From C23-C27, the nonlinearity effect becomes more significant because SVOC vapour pressures  
534 decrease with carbon number making evaporation slower. For those SVOCs having a timescale of  
535 condensation/evaporation comparable to the dilution timescale, the effects of both  
536 condensation/evaporation and neighbourhood dilution are significant, leading to significant  
537 nonlinearity.

538

539 For higher carbon components (C28-C32), vapour pressures are very small (Table 1) and emission  
540 rates are relatively smaller than lower carbon components, so only rather slow  
541 condensation/evaporation processes occur. Their nonlinearities are less significant, with the dilution  
542 process dominating, and only a slight influence of condensation/evaporation.

543

544 The sum of gas and particles (Figure 6(c)) can be scaled well although either gas or particles for  
545 higher carbon components cannot be scaled well, indicating the evolving partitioning between gas  
546 and particles. Figure 6(d) shows correction factors for a selected nonlinear carbon SVOC (C25) at  
547 the downwind location of rooftop+400m in cases with varying background and emissions. These  
548 correction factors can be up to around 60% for higher background cases and lower emission cases. A  
549 consistent pattern is that, as  $\alpha$  (or  $\beta$ ) increases, the gas concentration of C25 decreases; in other words,  
550 increasing  $\alpha$  (or  $\beta$ ) will suppress the evaporation of SVOCs on the particles. Correction factors for  
551 gas concentrations always have opposite signs, and occur in a certain ratio to those for particle  
552 concentrations, indicating that the conversion between gas and particles is conservative (Figure 6(c)).

553 This suggests a relationship of correction factors between gas and particles, i.e.  $\phi_{gas}^{ic} / \phi_{particle}^{ic} =$   
554  $-\hat{C}_{particle}^{BASE} / \hat{C}_{gas}^{BASE}$ , which can also be inferred from Equations 7-8 assuming the conservation of the  
555 sum of gas and particle concentrations ( $\hat{C}_{gas}^{ic} + \hat{C}_{particle}^{ic} = \hat{C}_{gas}^{BASE} + \hat{C}_{particle}^{BASE}$ ). The asymmetric  
556 patterns of correction factors between the gas phase and the particle phase in Figure 6(d) are due to  
557 the BASE value for gas being lower than that for particles.



559 **4. CONCLUSIONS**

560 Size-dependent multicomponent microphysics of UFPs (involving condensation/evaporation of  
561 SVOCs) has been coupled with WRF-LES to simulate the neighbourhood dispersion of UFPs for an  
562 idealised line (street) emission. UFPs are horizontally advected downwind into the neighbourhood  
563 scale and vertically mixed with urban background air. There is evidence of evaporation effects, which  
564 alter the size distribution of UFPs. The lightest SVOCs evaporate within a model time step of 0.2 s,  
565 while those in a middle range of carbon numbers (C21-C26) evaporate with a timescale about equal  
566 to that of mixing. The dispersion and evolution of UFPs at the neighbourhood-scale are the result of  
567 combined effects among emissions, mixing with background and evaporation/condensation.  
568 Among a wide range of timescales for the evaporation/condensation of SVOCs (Table S4 in the  
569 Supplementary Material), only those of the same order of magnitude as the travelling time have  
570 significant contributions to the non-linear part of the UFP concentration field during the  
571 neighbourhood-scale dispersion. There is a very good linearity for total UFPNC, UFPNSD (for UFP  
572 diameter greater than 100 nm), concentrations of lower carbon components, and concentrations of  
573 the sum of both gas and particles. The linearity is less precisely upheld for the number concentration  
574 of smaller particles and concentrations of those carbon components with a timescale comparable to  
575 the dilution timescale. For the non-perfectly scaled quantities, a linear model may be adopted to yield  
576 an approximate solution with a tolerance of an error. For the value range of  $\alpha$  and  $\beta$  tested in this  
577 study, the errors are about 10% or less for the nucleation mode of particle number concentrations, but  
578 can be as large as around 60% for the mass concentrations of medium-carbon SVOC components.

579

580 The WRF-LES-UFP model developed in this study reveals the evolution and dispersion of sized-  
581 resolved UFPs from an urban street to its neighbourhood-scale surroundings and can be extended  
582 straightforwardly to simulate the neighbourhood scale dispersion of UFPs for a realistic street canyon  
583 network. Gridded surface emissions based on the real-time traffic reactivity can be implemented as

584 an input in the model to represent a real-world street network. Planned future modelling work is to  
585 configure the model with a realistic street canyon network emission pattern and to conduct sensitivity  
586 tests of key parameters under different atmospheric conditions.

587

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595

## 596 **CODE AVAILABILITY**

597 WRF v3.6.1 is available at [http://www2.mmm.ucar.edu/wrf/users/download/get\\_sources.html](http://www2.mmm.ucar.edu/wrf/users/download/get_sources.html). The  
598 coupling WRF v3.6.1 large eddy simulation code with UFP microphysics module and case settings  
599 are archived on Zenodo (<https://doi.org/10.5281/zenodo.3333811>).

## 600 **DATA AVAILABILITY**

601 Data supporting this publication are openly available from the UBIRA eData repository at  
602 <https://doi.org/10.25500/edata.bham.00000366>

603

604

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## TABLE LEGEND:

**Table 1.** Initial/background partial pressure (derived from vapour concentrations on a urban background site) and saturation vapour pressures estimated at temperature of 284.15 K based on the UManSysProp online tool using data from Compernolle et al. (2011).

## FIGURE LEGENDS:

**Figure 1.** Total UFP number concentration ( $\# \text{ cm}^{-3}$ ): Vertical slice of (a) mean concentration ( $C_m$ ) and (b) the ratio of concentration fluctuation intensity to mean concentration ( $C'/C_m$ ); (c) mean concentration with fluctuations along South-North line (starting from 200 m before the emission) at the bottom level.

**Figure 2.** (a) UFP number-size distribution  $dN/d\log D_p$  ( $\# \text{ cm}^{-3}$ ) (fluctuation intensity indicated by shaded areas) for rooftop and its downwind neighbourhood at the bottom level; The inflowing background is taken from 200 m before the emission at the bottom level of the domain; The size-dependent emission flux  $dE/d\log D_p$  is shown for comparison. (b) Dilution ratio for the downwind locations; Dashed lines represent the dilution ratio for a passive-like scalar (e.g. total UFP number concentration) at each downwind locations. Rooftop +100 m and +400 m represent the travelling time of about 50 m and 200 s, respectively, in this study.

**Figure 3.** SVOCs concentration ( $\text{ng m}^{-3}$ ) (fluctuation information indicated by shaded areas) at the rooftop and its downwind neighbourhood locations at the bottom level: (a) Gas concentrations; (b) Particle concentrations, (c) Sum of gas and particle concentrations and (d) Dilution ratio for the downwind locations (The absolute concentrations for high-carbon gas-phase components and for low-carbon particle-phase components are very close to the background concentrations; therefore the dilution ratios for these components are not shown.); Dash lines represent the dilution ratio for a passive-like scalar (also indicated in Figure 4b) at each downwind locations. Rooftop +100 m and +400 m represent the travelling time of about 50 m and 200 s, respectively, in this study.

**Figure 4.** Scaled total UFP number concentration along South-North line at the bottom level.

**Figure 5.** (a) Scaled UFP number-size distributions and (b)  $\phi$  for selected bins (bins 3, 5 and 7 with bin bounds of [10.7 nm, 14.6 nm], [19.8 nm, 26.9 nm] and [36.6 nm, 49.9 nm], respectively) at rooftop+400m (or travelling time of about 200 s) for the bottom level.

**Figure 6.** Scaled SVOC (a) gas concentrations, (b) particle concentrations, (c) the sum of both gas and particle concentrations and (d) Correction factor for a typical SVOC C25 for rooftop+400m (or travelling time of about 200 s) at the bottom level.

845 **Table 1.** Initial/background partial pressure (derived from vapour concentrations on an urban  
846 background site) and saturation vapour pressures estimated at temperature of 284.15 K based on the  
847 UManSysProp online tool using data from Compennolle et al. (2011).

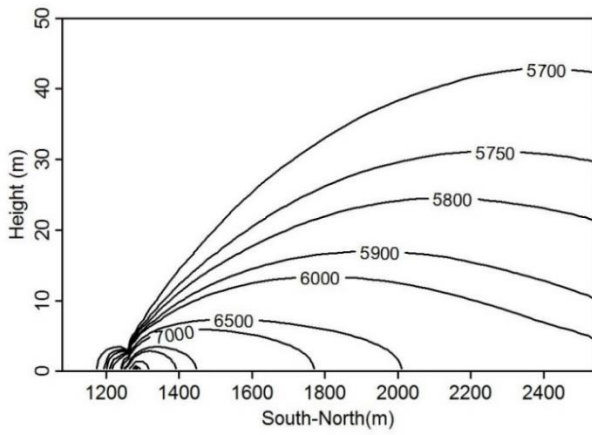
N-alkanes	Partial pressure (in Pa)	Saturation vapour pressure at 284.15 K (in Pa)
C16	1.19E-007	6.42E-02
C17	8.90E-008	1.91E-02
C18	4.44E-008	5.69E-03
C19	2.77E-008	1.70E-03
C20	1.20E-008	5.05E-04
C21	4.16E-009	1.50E-04
C22	1.29E-008	4.48E-05
C23	1.73E-008	1.33E-05
C24	2.29E-008	3.97E-06
C25	1.76E-008	1.18E-06
C26	8.74E-009	3.52E-07
C27	5.73E-009	1.05E-07
C28	5.80E-009	3.12E-08
C29	3.57E-009	9.30E-09
C30	2.73E-009	2.77E-09
C31	4.46E-009	8.25E-10
C32	4.31E-009	2.46E-10

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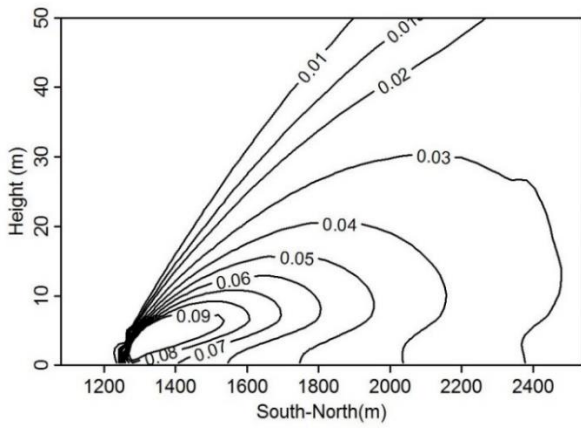


850 (a)  $C_m$  (# cm<sup>-3</sup>)



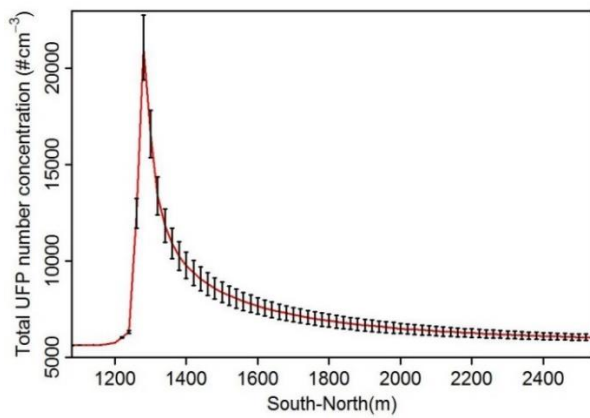
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852 (b)  $C'/C_m$



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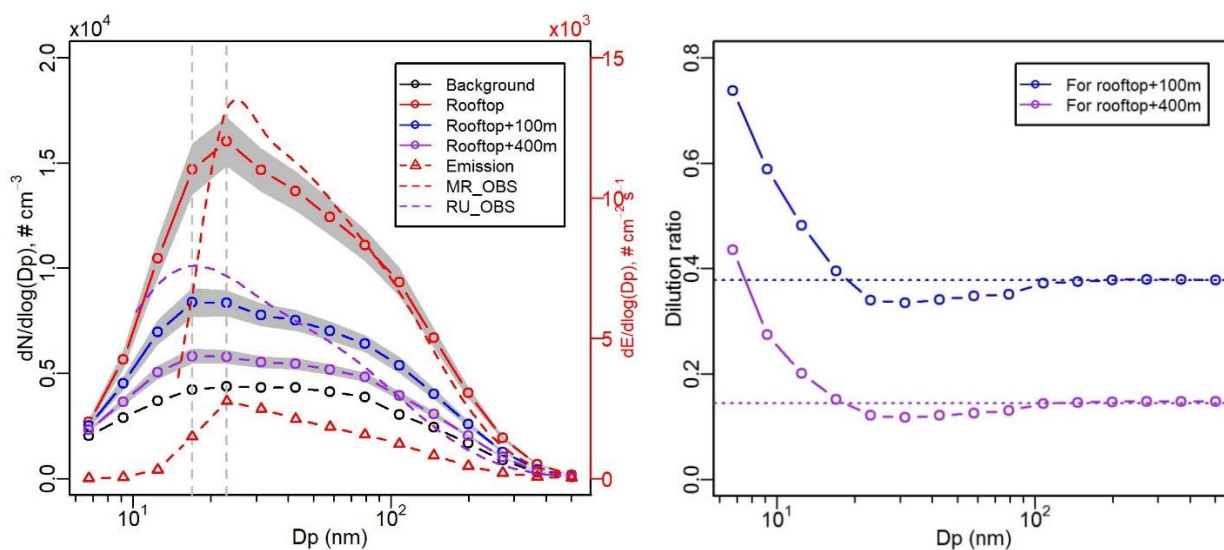
854 (c)  $C_m \pm C'$  along the South-North line



855

856 **Figure 1.** Total UFP number concentration (# cm<sup>-3</sup>): Vertical slice of (a) mean concentration ( $C_m$ )  
 857 and (b) the ratio of concentration fluctuation intensity to mean concentration ( $C'/C_m$ ); (c) mean  
 858 concentration with fluctuations along South-North line (starting from 200 m before the emission) at  
 859 the bottom level from the model.

860 (a) (b)

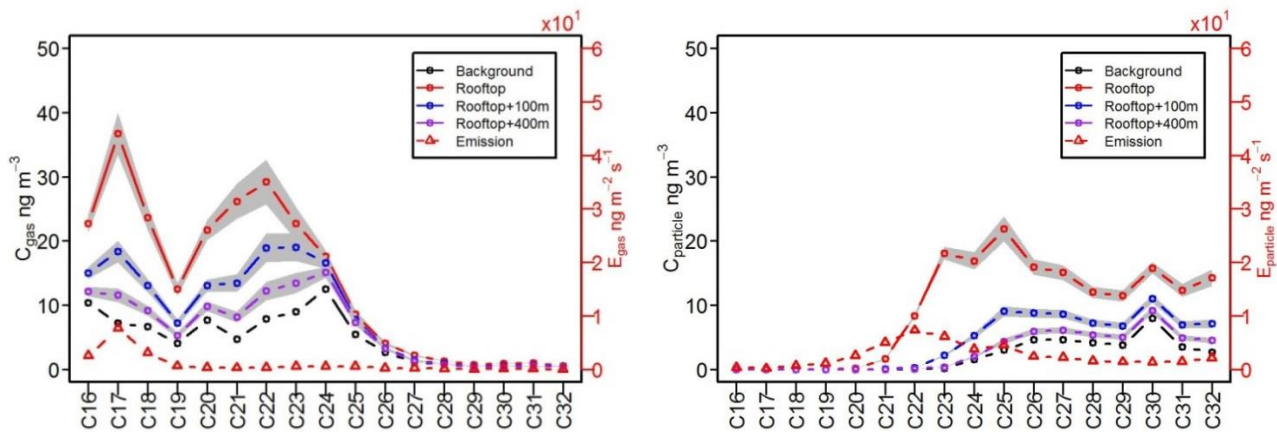


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862 **Figure 2.** (a) UFP number-size distribution  $dN/d\log D_p$  ( $\# \text{ cm}^{-3}$ ) (fluctuation intensity indicated by  
863 shaded areas) for rooftop and its downwind neighbourhood at the bottom level; The inflowing  
864 background is taken from 200 m before the emission at the bottom level of the domain; The size-  
865 dependent emission flux  $dE/d\log D_p$  is shown for comparison; MR\_OBS represents the measurement  
866 from Marylebone roadside; RU\_OBS represents the measurement from the nearby downwind rooftop  
867 location. (b) Dilution ratio for the downwind locations; Dashed lines represent the dilution ratio for a  
868 passive-like scalar (e.g. total UFP number concentration) at each downwind locations. Rooftop +100  
869 m and +400 m represent the travelling time of about 50 m and 200 s, respectively, in this modelling  
870 study.

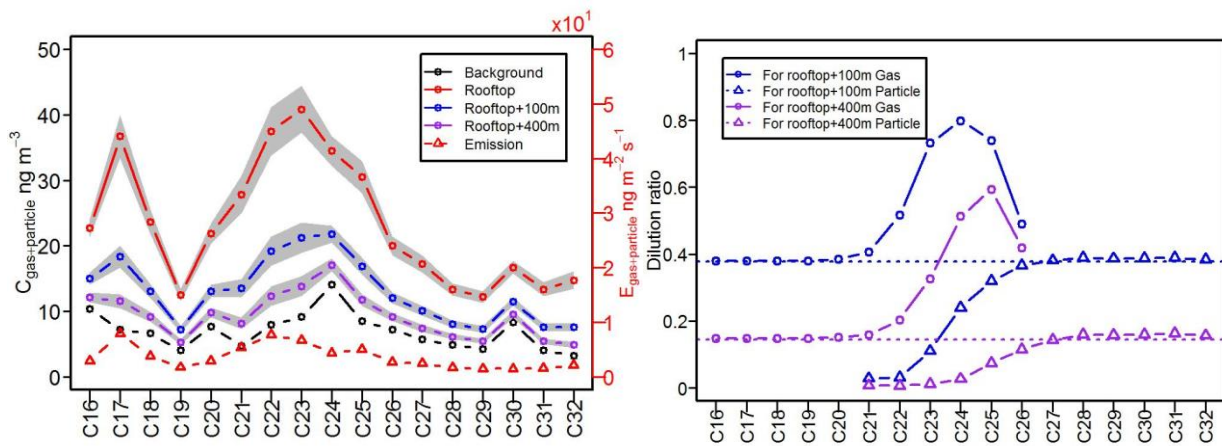
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872 (a) (b)



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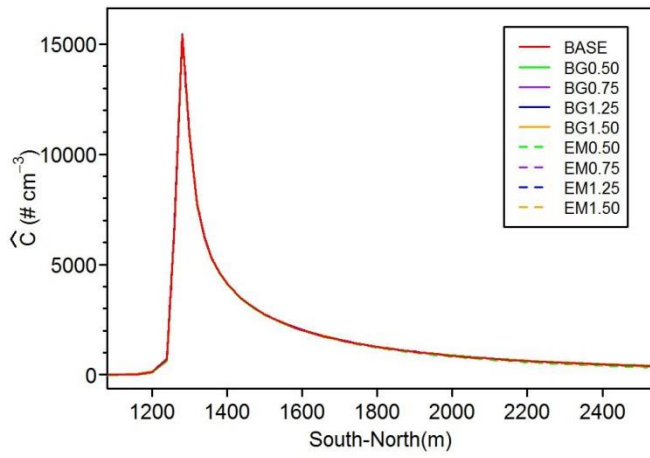
874 (c) (d)



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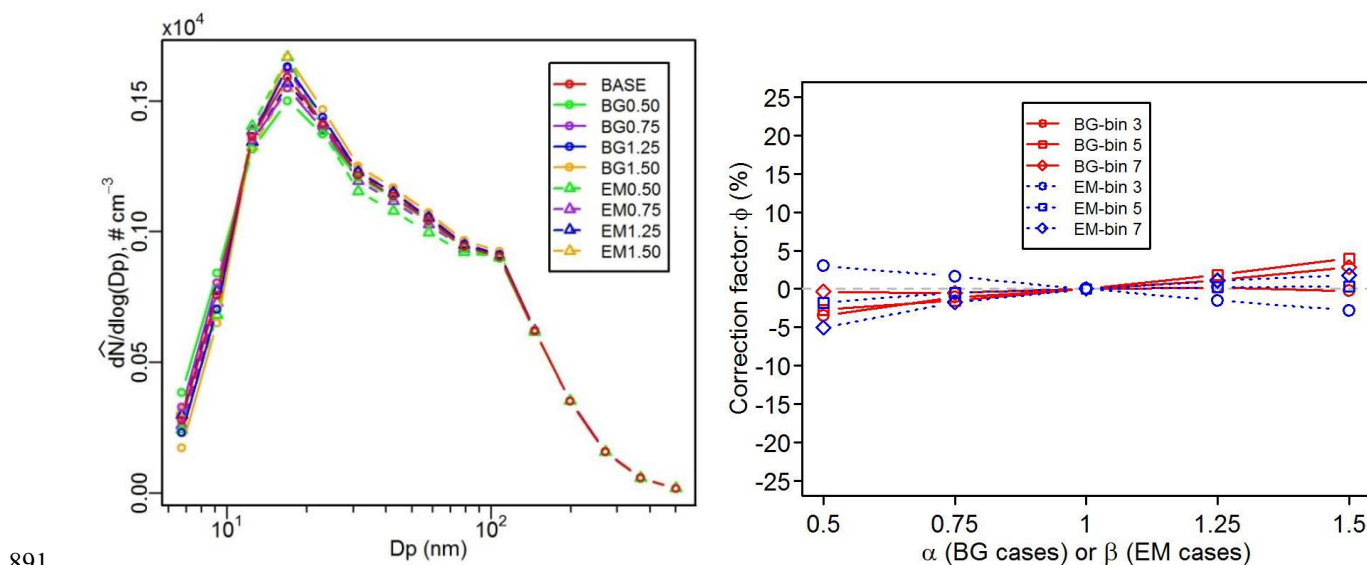
876 **Figure 3.** SVOCs concentration (ng m<sup>-3</sup>) (fluctuation information indicated by shaded areas) at the  
877 rooftop and its downwind neighbourhood locations at the bottom level: (a) Gas concentrations; (b)  
878 Particle concentrations, (c) Sum of gas and particle concentrations and (d) Dilution ratio for the  
879 downwind locations (The absolute concentrations for high-carbon gas-phase components and for  
880 low-carbon particle-phase components are very close to the background concentrations; therefore the  
881 dilution ratios for these components are not shown.); Dash lines represent the dilution ratio for a  
882 passive-like scalar (also indicated in Figure 4b) at each downwind locations. Rooftop +100 m and  
883 +400 m represent the travelling time of about 50 m and 200 s, respectively, in this modelling study.

884



**Figure 4.** Scaled total UFP number concentration along South-North line at the bottom level from the model.

890 (a)  $d\hat{N}/d\log(D_p)$  at rooftop+400m (b)  $\phi$  for selected bins at rooftop+400m

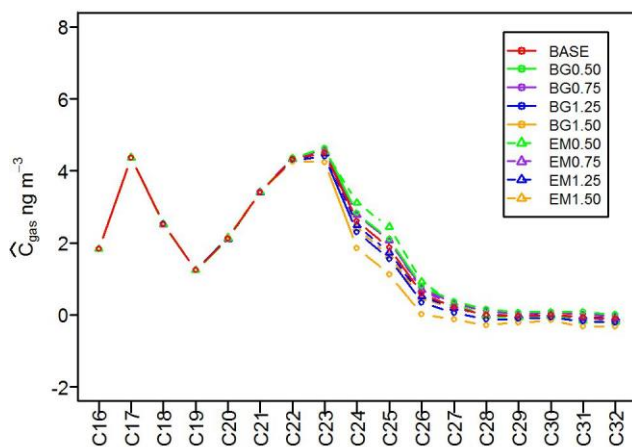


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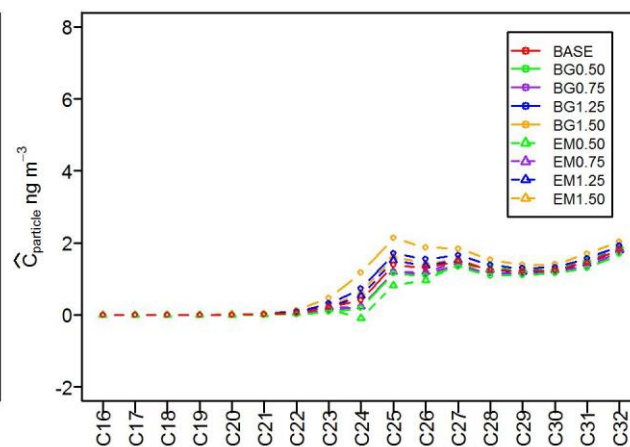
892 **Figure 5.** (a) Scaled UFP number-size distributions and (b)  $\phi$  for selected bins (bins 3, 5 and 7 with  
893 bin bounds of [10.7 nm, 14.6 nm], [19.8 nm, 26.9 nm] and [36.6 nm, 49.9 nm], respectively) at  
894 rooftop+400m (or travelling time of about 200 s) for the bottom level from the model.

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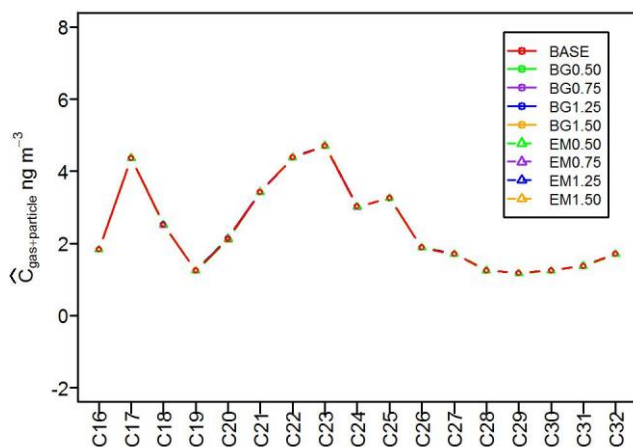
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897 (a)  $\hat{C}_{gas}$  at rooftop+400m

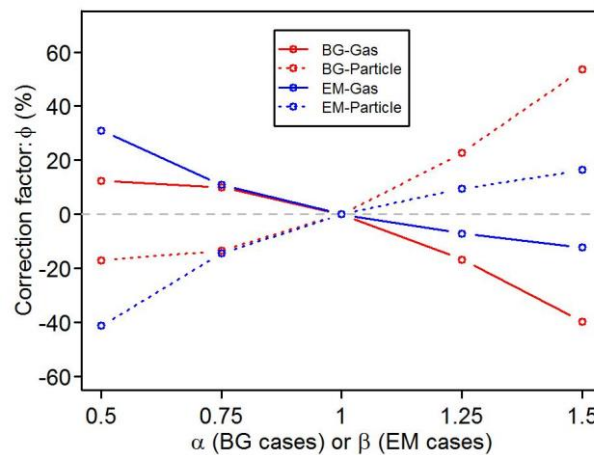
898

(b)  $\hat{C}_{particle}$  at rooftop+400m

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900 (c)  $\hat{C}_{gas+particle}$  at rooftop+400m

901

(d)  $\phi$  for C25 at rooftop+400m

902 **Figure 6.** Scaled SVOC (a) gas concentrations, (b) particle concentrations, (c) the sum of both gas  
 903 and particle concentrations and (d) Correction factor for a typical SVOC C25 for rooftop+400m (or  
 904 travelling time of about 200 s) at the bottom level from the model.

905

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907